## Experimental observation of shear thickening oscillation

Shin-ichiro Nagahiro

Department of Mechanical Engineering, Sendai National College of Technology, Natori, Miyagi 981-1239, Japan

## Hiizu Nakanishi

Department of Physics, Kyushu University 33, Fukuoka 812-8581, Japan

## Namiko Mitarai

Niels Bohr Institute, University of Copenhagen, Blegdamsvej 17, DK-2100, Copenhagen Ø, Denmark (Dated: November 14, 2012)

We report experimental observation of the shear thickening oscillation, i.e. the spontaneous macroscopic oscillation in the shear flow of severe shear thickening fluid. The shear thickening oscillation is caused by the interplay between the fluid dynamics and the shear thickening, and has been predicted theoretically by the present authors using a phenomenological fluid dynamics model for the dilatant fluid, but never been reported experimentally. Using a density-matched starch-water mixture, in the cylindrical shear flow of a few centimeters flow width, we observed strong vibrations of the frequency around 20 Hz, which is consistent with our theoretical prediction.

PACS numbers: 83.80.Hj,83.60.Rs, 83.10.Ff,83.60.Wc

Introduction: Fluid media such as dense colloids or dense granule-fluid mixtures are often called dilatant fluids and known to show severe shear thickening, i.e. its viscosity changes discontinuously by orders of magnitude upon increasing the shear rate[1–4]. This is a source of a variety of unintuitive curious behaviors of the media[5–7], and might be used for interesting application like a body armor[8], but often causes problems in industrial situations, such as the damage of mixer motors due to overloading[1].

Although the severe shear thickening is a property that can be demonstrated easily with common material like starch and water, physicists have not reached a general agreement on its microscopic mechanism. Originally, the order-disorder transition of the dispersed particles was proposed[1, 9–11], but later the hydrodynamics cluster formation by the lubrication force is considered to be more consistent with experimental observations and numerical simulations[12–15]. More recently, the transition is discussed in connection with the jamming and/or the compaction of granules[16–19].

Recently, the present authors constructed a fluid dynamics model for the dilatant fluid and found that, in a certain shear flow situations, the medium should show a spontaneous oscillation, which we call the *shear thick-ening oscillation*[20, 21]. Our model is based upon fluid dynamics with a phenomenological field  $\phi(\mathbf{r})$  representing an internal state of the medium. The internal state  $\phi(\mathbf{r})$  is determined by the local stress, and the viscosity of the medium is a function of  $\phi(\mathbf{r})$ . By assuming simple functional relations among these quantities, we have demonstrated that the model shows basic properties of the dilatant fluids, such as a discontinuous increase and hysteresis in the viscosity upon changing the shear rate.

The shear thickening oscillation that the model predicts for the fluid in the shear flow is a salient property, but has not been reported in the literature. When the ex-

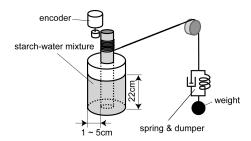


FIG. 1: Schematic illustration of the experimental setup consisting of a cylindrical container, a rotating rod, and a rotary encoder.

ternal stress is in a certain range, the internal state of the medium should alternate between a thickened state and a relaxed thin state due to the interplay between the fluid dynamics and the internal state dynamics. Such a hydrodynamics oscillation of the medium has neither been predicted theoretically nor been reported experimentally. By estimating the parameters in the model using experimental data for the cornstarch-water mixture [2], the oscillation frequency is expected to be of the order of 10 Hz when the flow width is several centimeters, thus it is really a hydrodynamic oscillation, not due to a discreteness of granules. It should be noted that the oscillation is suppressed by the viscosity when the flow width is of the order of millimeters, i.e. a typical size of rheometer sample for precision measurement; This may be the reason why the shear thickening oscillation has not been reported yet. In this paper, we report experimental observation of spontaneous oscillation in the cylindrical Couette flow of the starch-water mixture.

Experimental Setup: Figure 1 illustrates our experimental setup. The fluid flows in the Taylor-Couette geometry between the outer cylinder and the coaxial rod at the center. The cylindrical container is 24 cm tall,

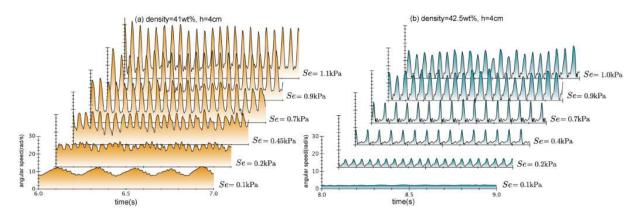


FIG. 2: (Color online) Time evolutions of angular speed of the rod for the flow of h=4 cm thickness with various applied stresses. Suspension densities are (a) 41 wt% and (b) 42.5 wt%.

but is filled with the fluid upto 22 cm deep. The diameter of the center rod is 5 cm, while we use several outer cylinders with different inner diameters, so that we can have the flow of the width  $h=1\sim 5$  cm thicknesses. The outer cylinder and the rod are both acrylic, and their surfaces are lined with water-proof sand paper in order to enforce the no-slip boundary condition. The center rod rotates under an externally applied constant torque with the outer cylinder being fixed. The external torque is applied by a weight through a steel wire wound on the rod; the weight is hung through a spring and dumper system (Samini Co., Ltd.), and its weight is in the range of  $0.5 \sim 10$  kg, which gives the external stress  $S_e = 0.14 \sim 2.8$  kPa at the surface of the rod. The angular speed of the center rod  $\omega$  is recorded by a rotary encoder (SP-405ZA, Ono Sokki). As for the fluid, we use the suspension of potatostarch (Hokuren) densitymatched with the aqueous solution of CsCl of the density 1.6 g/cm<sup>3</sup>. The upper surface of the fluid is open to the air although the container is capped with an aluminum plate.

Shear Thickening Oscillation: Figure 2 shows the time evolutions of the angular speed of the rod for the potatostarch suspension of the concentrations 41 wt% (a) and 42.5 wt% (b) with the flow width h = 4 cm. The plots show the oscillations for the various external shear stresses  $S_e$  at the surface of the rod. For both of the concentrations, clear oscillations about 20 Hz are observed for the external stress  $S_e \gtrsim$  0.2 kPa. The shape of the oscillations for the 41 wt% suspension is rather symmetric and sinusoidal, while for the 42.5 wt% suspension the oscillation is nearly symmetric at  $S_e = 0.2$  kPa, but becomes more asymmetric for larger  $S_e$ , i.e. a sawtooth oscillation that consists of a gradual increase of the angular speed followed by a sudden drop. For the small external stress of  $S_e = 0.1$  kPa, the 20 Hz oscillation disappears, but a slower oscillation around 5 Hz is observed in the case of the thinner fluid of the 41 wt% concentration (Fig.2(a)). For the large external stress, we expect that the rod will be stuck due to the severe shear thickening, but in the present experiment, for  $S_e \gtrsim 2.0$  kPa,

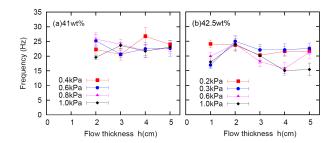


FIG. 3: (Color online) The frequency of the shear thickening oscillation for (a) 41 wt% and (b) 42.5 wt% potatostarch suspension as a function of flow thickness h. The plots are evaluated from the average interval of neighboring peaks, and the errorbars represent standard deviation.

the rod starts slipping suddenly after initial transient.

Figure 3 shows the oscillation frequency as a function of the flow width h for various external stress  $S_e$ . One can see that the frequencies are around 20 Hz for all the cases, and we could not find any systematic dependence on neither h nor  $S_e$ . For  $S_e$  higher than 1.0 kPa, the center rod starts slipping and we could not obtain clean data, but we find no clear sign of a systematic change in the frequency upto  $S_e = 2.3$  kPa.

Figure 4 shows the maximum and minimum angular speed of the rod,  $\omega_{\rm max}$  and  $\omega_{\rm min}$ , during a single cycle of oscillation against  $S_e$  for some values of the flow width h for both of the concentrations. Each data point represents an average over a single run of the experiment, which contains typically  $10^2$  cycles of the oscillation. It should be notable that both  $\omega_{\rm max}$  and  $\omega_{\rm min}$  barely depend on the flow width h. As for the maximum angular speed  $\omega_{\rm max}$ , the plots show almost linear dependence on the external shear stress  $S_e[22]$ . On the other hand, the minimum angular speed  $\omega_{\min}$  stays roughly constant for  $S_e \gtrsim 0.5$  kPa after the initial decrease for smaller  $S_e$ . In the case of the 42.5 wt% suspension, the oscillation disappears around  $S_e \lesssim 0.2$  kPa, for which cases the terminal angular speeds are plotted with open marks in Fig.4(c) and (d).

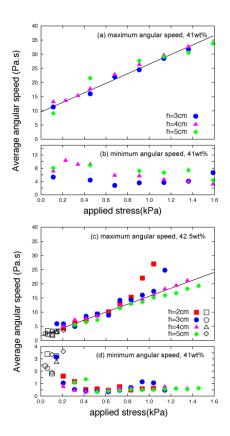


FIG. 4: (Color online) The maximum and minimum angular speed of the center rod during the oscillations as a function of the applied stress  $S_e$  for (a,b) 41 wt% and (c,d) 42.5 wt% potatostarch suspensions. Each data point represents an average over a single run of experiment, which contains typically  $10^2$  cycles of the oscillation. The open symbols in (c,d) represent the angular speed of the non-oscillating flows for small  $S_e$ .

The linear dependence of  $\omega_{\rm max}$  on  $S_e$  leads us to introduce an average viscosity  $\eta_{\rm relax}$  for the flow during the relaxed state. If we assume the steady cylindrical Couette flow with a constant angular velocity  $\omega$ , the viscosity would be given by

$$\eta = \frac{S_e}{2\omega} \left[ 1 - \left( \frac{R_1}{R_2} \right)^2 \right],\tag{1}$$

where  $R_1=2.5$  cm is the radius of the center rod and  $R_2=R_1+h$  is the radius of the outer cylinder. For the 42.5wt% suspension in the relaxed state, the plots in Fig.4(c) shows  $\omega_{\rm max}\approx 15S_e$  rad/s. Substituting this  $\omega_{\rm max}$  into  $\omega$  in Eq.(1), we have  $\eta_{\rm relax}=20\sim 30$  Pa·s, which is consistent with the previous viscosity measurement before the thickening transition [2]. For the 41 wt% suspension, Fig.4(a) shows  $\omega_{\rm max}\approx 10+11S_e$  rad/s, which has a substantial non-zero extrapolation at  $S_e=0$ . This may be due to the inertia effect in the oscillatory state and our steady state formula, Eq.(1), is not valid for this case.

In the thickened state,  $\omega_{\min}$  is nearly constant around

 $0.5~{\rm rad/s}$  for  $0.5\lesssim S_e\lesssim 1.5~{\rm kPa}$  for the 42.5wt% suspension, thus the viscosity in the thickened state may be estimated to be of the order of  $10^3~{\rm Pa\cdot s}$ , which seems to be somewhat smaller than the value observed in Ref.[2]. This may suggest that the whole system is not thickened uniformly, but the thickening region is localized as we will discuss later.

Comparison with model behaviors: Some features we observed are consistent with those predicted by our phenomenological model[21]. The shear flow starts oscillating under a constant shear stress larger than a certain value. The oscillation is sinusoidal for the stress near the threshold, but becomes asymmetric for larger stress in the sawtooth shape. The observed frequency of the oscillation is about 20 Hz, which is roughly in the range expected by the model for the flow width of a several centimeters.

On the other hand, there are some other features that seem difficult to interpret in terms of the simple shear flow oscillation. The oscillation frequency does not depend significantly neither on the external stress  $S_e$  nor on the flow width h, whereas the simulations of cylindrically symmetric oscillation show that the frequency tends to become slightly higher as  $S_e$  increases, and that the frequency decreases significantly as the flow width h increases. It is also tricky to interpret the behavior of the maximum  $\omega_{\rm max}$  and minimum angular speed  $\omega_{\rm min}$  of the oscillation:  $\omega_{\rm max}$  is proportional to  $S_e$  while  $\omega_{\rm min}$  stays almost constant, and neither of them do not depend on the flow width h. We find that these features are not easy to reproduce by the model as long as we assume that the flow is cylindrically symmetric.

In fact, the simulations in two dimensions show that a localized thickened band region appears when the cylindrical Poiseuille flow is destabilized beyond a certain  $S_e$ . Figure 5 shows the results of numerical simulations of the fluid dynamics model[20, 21] for the time developments of the flow when the inner cylinder is driven toward CCW with the outer cylinder being fixed. The velocity field  $\boldsymbol{v}(\boldsymbol{r})$  and the spatial variation of the viscosity  $\eta(\boldsymbol{r})$  are shown by the arrows and the gray scale maps, respectively.

The initial destabilization process of the cylindrically symmetric flow is shown in Fig.5(a) after the system starts from the static state. As the inner cylinder starts rotating, a uniform circular flow is induced and the region near the inner cylinder thickens, then a cylindrically symmetric state is destabilized. A localized thickened region forms a band structure to extend in the radial direction, then the flow slows down drastically when the thickening band reaches the outer cylinder.

After the initial transient, the system undergoes steady periodic oscillation, where the thickening band breaks off and extends periodically during the course of the oscillation (Fig.5(b)). The band goes around with the inner cylinder. Our tentative results of the simulations show that, for this oscillatory flow with a thickening band, the frequency is determined by the way how the band

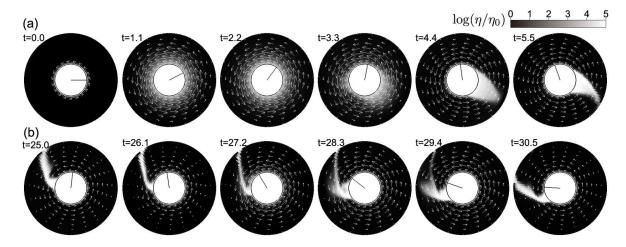


FIG. 5: Time developments of cylindrical shear flows by the simulations of the fluid dynamics model of shear thickening[21]:
(a) the initial destabilization of the cylindrically symmetric flow from the static state, (b) one cycle of steady oscillation with a localized thickening band. The inner cylinder is driven toward CCW by the external shear stress  $S_e$  with the outer cylinder being fixed. The rotation of the inner cylinder is represented by the solid lines at the center. The velocity field v(r) and the spatial variation of the viscosity  $\eta(r)$  relative to that in the relaxed state  $\eta_0$  are shown by the arrows and the grey scale maps, respectively. The simulation parameters are the driving shear stress  $S_e = 3$ , the radius of inner cylinder  $R_1 = 1$ , and the flow width h = 2 in the dimensionless unit system defined in Ref.[21]. The model parameters are r = 0.1, A = 1 and  $\phi_M = 0.85$ . The moment of inertia of the inner cylinder is neglected.

breaks off, and its dependence on the flow width h and the shear stress  $S_e$  is weak in comparison with the case of the cylindrically symmetric flow. This may explain our experimental data. Detailed behavior of the oscillatory flow with this localized thickening band is now under investigation.

In summary, we observed the shear thickening oscillation around 20 Hz in the shear flow of the Taylor-Couette geometry with several centimeter flow width as has been predicted theoretically. The frequency and the amplitude

of the oscillation barely depend on the flow thickness. Numerical simulations in two dimensions suggests that the oscillation occurs with a thickening band that breaks off and extends periodically.

We thank T. Kato, T. Sugawara, and Y. Fukuda for technical assistance. This work is supported by KAK-ENHI grant number 21540418 (H.N.) and 24760145 (S. N.), and the Danish Council for Independent Research, Natural Sciences (FNU) (N.M.).

- [1] H. Barnes, J. Rheology 33, 329 (1989).
- [2] A. Fall, N. Huang, F. Bertrand, G. Ovarlez, and D. Bonn, Phys. Rev. Lett. 100, 018301 (2008).
- [3] E. Brown and H. M. Jaeger, Phys. Rev. Lett. 103, 086001 (2009).
- [4] N. J. Wagner and J. F. Brady, Physics Today 62(10), 27 (2009).
- [5] F. S. Merkt, R. D. Deegan, D. I. Goldman, E. C. Rericha, and H. L. Swinney, Phys. Rev. Lett. 92, 184501 (2004).
- [6] H. Ebata, S. Tatsumi, and M. Sano, Phys. Rev. E 79, 066308 (2009).
- [7] S. von Kann, J. Snoeijer, D. Lohse, and D. van der Meer, Phys. Rev. E(R) 84, 060401 (2011).
- [8] Y. S. Lee, E. D. Wetzel, and N. J. Wagner, J. Mater. Sci. 38, 2825 (2003).
- [9] R. Hoffman, Trans. Soc. Rheol 16, 155 (1972).
- [10] R. Hoffman, J. Colloid Interface Sci. 46, 491 (1974).
- [11] R. Hoffman, J. Rheol. 42, 111 (1998).
- [12] J. Bender and N. J. Wagner, J. Rheol. 40, 899 (1996).
- [13] B. J. Maranzano and N. J. Wagner, J. Chem. Phys. 117,

- 10291 (2002).
- [14] J. Brady and G. Bossis, J. Fluid Mech. 155, 105 (1985).
- [15] J. Melrose and R. Ball, J. Rheol. 48, 937 (2004).
- [16] D. Lootens, H. van Damme, Y. Hémar, and P. Hébraud, Phys. Rev. Lett. 95, 268302 (2005).
- [17] E. Brown et al., Phys. Rev. E 84, 031408 (2011).
- [18] E. Bertrand, J. Bibette, and V. Schmitt, Phys. Rev. E 66, 060401(R) (2002).
- [19] S. R. Waitukaitis and H. M. Jaeger, Nature 487, 205 (2012).
- [20] H. Nakanishi and M. Namiko, J. Phys. Soc. Jpn. 80, 033801 (2011).
- [21] H. Nakanishi, S. Nagahiro, and M. Namiko, Phys. Rev. E 85, 011401 (2012).
- [22] The three data pints for h=2 and 3 cm around  $S_e\approx 1.1$  kPa in Fig.4(a) are substantially off the general trend of the other data points. Judging from the time evolutions of  $\omega$ , these are due to intermittent slippage of the center rod, thus should be ignored in this analysis.